MAGNETIC CORRELATIONS AND HEAVY FERMION SYSTEMS

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Polarized and unpolarized neutron scattering measurements on both the Ce and U based heavy fermion systems have provided a great deal of evidence for the formation of a coherent state involving the f-electrons at low temperature. These experiments are reviewed, and common features of the low temperature magnetic correlations are discussed.

1. Introduction

As the list of heavy fermion systems (HFS) expands, and experiments delve deeper into the low temperature behavior of these materials, we seem to find more interesting new puzzles as well as clues to old questions. A great deal of insight into the microscopic description of the HFS has been provided by neutron scattering measurements. This technique is sensitive to the magnetic 4f(5f) electrons in the Ce(U) compounds, and so directly probes both the spatial and frequency response of the electrons which are at the heart of the physics of HFS.

Much of the attention of the field over the past year has been focussed on the correlations between the f-electrons that develop at low temperature. These correlations seem to be the feature which separates the HFS from the single ion Kondo systems, and have been explored theoretically in several papers [1-4]. Additionally, the HFS are characterized by a delicate balance between interactions which favor different ground states (eg. superconductivity, magnetic order, paramagnetism). One therefore finds extremely complicated low temperature behavior which, for example, produces both an antiferromagnetically ordered ground state and superconductivity in URu_2Si_2 and UPt_3, along with additional spin-fluctuation modes in UPt_3 of an entirely different character.

In this review, I will concentrate on the experimental evidence, from neutron scattering measurements, for magnetic correlations at low temperatures in some Ce and U-based HFS. Therefore, it is useful to first briefly summarize what is measured by this technique. The energy and $\vec{q}$-dependent cross-section for magnetic scattering is,

$$\frac{d^2\sigma}{d\omega d\Omega} = \gamma_0^2 \left(\frac{g_j}{2}\right)^2 \frac{k_f}{k_i} f^2(Q) S(\vec{q},\omega),$$

where the symbols have their ususal meaning [5]. The quantity of interest here is the dynamical structure factor, $S(\vec{q},\omega)$, where $\vec{q}$ is measured from some chosen zone center in the reciprocal lattice along a specific direction. In addition to the non-trivial $\vec{q}$-dependence of $S(\vec{q},\omega)$ in the presence of magnetic correlations, the magnetic cross-section falls off monotonically with the magnetic form factor $f(Q)$. $S(\vec{q},\omega)$ in turn is related to the magnetic response, $\chi''(\vec{q},\omega)$ by

$$S(\vec{q},\omega) = \left(1 - \exp(\frac{\hbar\omega}{kT})\right)^{-1} \frac{\chi''(\vec{q},\omega)}{g_j^2 \mu_B^2 \pi}.$$

For most cases, in the absence of crystal field excitations, the frequency dependence of $\chi''(\vec{q},\omega)$ is well described by a quasielastic lorentzian of half-width Γ_q,

$$\chi''(\vec{q},\omega) = \chi(\vec{q}) \frac{\Gamma_q}{\Gamma_q^2 + \omega^2}.$$

Here, the subscript q means that this energy scale may have some non-trivial $\vec{q}$-dependence itself.

For the discussion that follows, it is important to realize that in a polycrystalline measurement we obtain only powder averaged (Brillouin zone averaged) information, so much of the $\vec{q}$-dependence of $S(\vec{q},\omega)$ is obscured, and the energy scale of the magnetic excitations is also averaged. Hence for polycrystalline samples, $\Gamma_q \rightarrow \Gamma$. In light of these remarks, the identification of magnetic correlations from powder data is quite difficult. Measurements on single crystal samples are generally required.

In the next two sections, several examples of neutron measurements on both Ce and U-based HFS are reviewed. In the last section there is some discussion of commonly observed features and suggestions for further study.

2. Ce-based HFS

2.1 $CeCu_6$

Neutron scattering measurements on $CeCu_6$ have been performed by several groups. Walter et al [6] studied the temperature dependence of the quasielastic linewidth from polycrystalline samples using the time-of-flight technique. Below 3K, the linewidth remains constant at Γ=0.5 meV, but grows ($\propto T^{\frac{1}{2}}$) as temperature increases. This $T^{\frac{1}{2}}$ dependence was also found in polycrystalline measurements on the HFS $CeAl_3$ by Murani et al [7] earlier.

Aeppli et al [8] investigated the $\vec{q}$-dependence and magnetic field dependence of the magnetic

scattering cross-section from single crystal specimens using unpolarized neutrons. In zero field, at low temperature, a maximum in the intensity of the magnetic scattering and a minimum in the quasielastic linewidth Γ_q was found at the (100) reciprocal lattice vector, where nuclear scattering is absent due to cancellation of the structure factor. A corresponding minimum in the magnetic intensity, and maximum in the linewidth was observed close to the nuclear (200) peak. This $\vec{q}$-dependence is consistent with the onset of antiferromagnetic correlations between the Ce^{3+} moments in the monoclinic unit cell below 10K [9]. Upon application of a magnetic field of 6.4T, the scattering is largely suppressed, confirming its magnetic origin.

Regnault et al [10] have also studied single crystals of $CeCu_6$ with unpolarized neutrons. In addition to confirming the presence of the antiferromagnetic spin fluctuations of reference 8, scans along the b* axis found no magnetic response. This indicates that the magnetic fluctuations are parallel to the monoclinic b-axis, in agreement with the Ising-like anisotropy seen in the magnetization measurements. In addition, the authors claim that inelastic measurements, performed at 0.1K, show evidence of a gap in the magnetic excitation spectrum of 0.25 meV which disappears at higher temperature.

2.2 $CeSi_xCu_y$

Very recently, two ferromagnetic HFS, $CeSi_{2-x}Cu_x$ [11] and $CeSi_{2-x}$ [12] have been studied. The onset of ferromagnetic order in $CeSi_{1.76}Cu_{0.24}$ (T_c = 8K) was investigated by Boni et al [13a] using unpolarized neutrons. They found that the ordered magnetic moment ($0.63 \pm 0.05\mu_B$) lies in the basal plane of the tetragonal structure. As seen also in the U based HFS, the ordered moment is significantly reduced from the paramagnetic moment of approximately $2.5\mu_B$.

One surprising feature of this investigation was the lack of any substantial critical scattering near the ordering temperature. In fact, only a small diffuse component was observed in the neighborhood of the transition in a double-axis experiment, while no critical scattering was found in a triple-axis measurements. As pointed out by Boni et al, this indicates that the spin fluctuations, even very close to T_c, are found at an energy scale larger than $k_BT_c \approx$ 1meV, the triple-axis energy resolution. In addition, no inelastic magnetic scattering could be found in the ordered phase.

Kohgi et al [13b] have performed inelastic neutron scattering measurements on $CeSi_{1.8}$. In this system, well developed magnetic excitations were observed below the ferromagnetic transition (T_c=13.4K). The linewidths of the excitations were rather large, and proportional to the excitation energy ($\Gamma_q \propto \omega_q$). The strong damping is perhaps due to interactions between the Ce f-electrons and the conduction electrons. It is possible that a further enhancement of this interaction in $CeCu_{0.24}Si_{1.76}$ broadens the magnetic excitations to the extent that they are unobservable. Clearly, further studies of these systems are called for.

3. U-based HFS

3.1 UPt_3

UPt_3 is perhaps the most thoroughly studied HFS by neutron scattering. Polycrystalline measurements using polarized neutrons by Aeppli et al [14] revealed a broad quasielastic response, Γ=10±2 meV, which essentially exhausts the bulk susceptibility at 1.3K. The measured fluctuating moment ($2.1 \pm 0.4\mu_B$) was consistent with that determined from the high temperature bulk susceptibility ($2.6 - 3.0\mu_B$). Single crystal measurements have revealed a great deal about the low temperature coherent state in this system, characterized by a decrease in the resistivity [15], anomalies in the Hall effect measurements [16], and a decrease in the in-plane bulk susceptibility below 18K [17]. There were also some surprises.

Single crystal unpolarized beam measurements [18] revealed the onset of antiferromagnetic correlations between the planes of U ions stacked along the c-axis of the hexagonal structure. Figure 1 shows a portion of the (*h0l*) plane of the reciprocal lattice of UPt_3. Maxima in the diffuse magnetic scattering are observed at the (001) and (003) reciprocal lattice points, with a corresponding minimum at (002). While nuclear reflections are found at wavevectors (0,0,2n), where the scattering from the atoms in the unit cell are in phase, the maxima at (0,0,2n+1) arise from short-range antiferromagnetic correlations between the two U ions in the unit cell. This modulation along the c-axis disappears upon warming the sample above ≈30K. Therefore, the onset of these antiferromagnetic spin fluctuations is in good accord with other bulk measurements indicating the onset of coherence at low temperature.

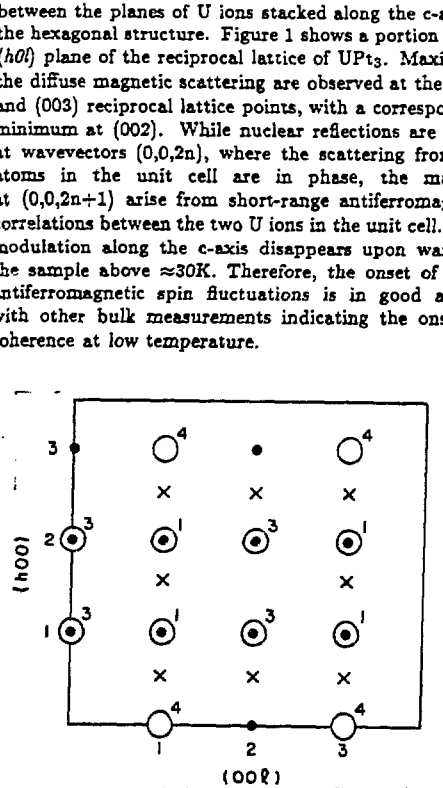

Figure 1. Section of the (*h0l*) reciprocal lattice plane in UPt_3. Solid circles represent the positions of the nuclear Bragg reflections. The open circles and numbers depict the positions and relative intensities of the maxima in the diffuse magnetic scattering (after ref. 20.). The crosses represent the positions of the magnetic Bragg reflections found in the diluted alloys (refs. 21 and 22).

Constant-Q scans taken at several points through the zone show that the energy scale of these spin fluctuations is $\Gamma_q \approx 5$ meV, close to the spin fluctuation temperature, T_{sf}, derived from specific heat measurements [19]. This value is somewhat smaller than, but not inconsistent with the previous polycrystalline measurements. Recall that the polycrystalline data is Brillouin-zone averaged, so that one actually measures an average spin fluctuation energy in this case. In addition, the energy scale of the spin fluctuations remains essentially constant across the Brillouin zone, and there is a finite, albeit smaller, cross-section at (002), in contrast to the expectations of a single-component, weakly interacting Fermi-liquid theory.

Subsequent polarized and unpolarized neutron scattering measurements [20] also explored the $\vec{q}$-dependence of the magnetic scattering transverse to the (00l) direction. A rather interesting feature of these data is the observation of a transverse modulation (eg. along (h01)), which persists to temperatures in excess of 100K, far above the onset of the antiferromagnetic correlations. This modulation arises from ferromagnetic correlations between U ions in the same basal plane, and is further enhanced by the strong antiferromagnetic coupling between planes below 30K.

In light of the magnetic correlations observed in UPt_3, it was very surprising to find magnetic ordering of a very different character in $U_{1-x}Th_xPt_3$ (x=0.03) [21] and $U(Pt_{1-y}Pd_y)_3$ (y=0.05) [22]. Both the magnetic structure and ordered moment ($0.65 \pm 0.1\mu_B$) for these alloys are the same. The magnetic unit cell is orthorhombic, obtained by doubling the hexagonal chemical unit cell along one of the in-plane axes, and the moment lies along this direction. The magnetic Bragg reflections are found at positions ($h/2,0,l$) in Fig. 1. The in-plane coupling is therefore antiferromagnetic, while the planes are coupled ferromagnetically!

Very recent high resolution measurements by Aeppli et al [23] indicates that pure UPt_3 does order antiferromagnetically at $T_N \approx 5$K with a structure which is identical to that of the diluted systems described above, but with a much smaller moment ($0.02 \pm 0.005\mu_B$). Several samples were studied in order to determine that the ordering was in fact intrinsic, and not due to defect or impurity effects. This is of course of some concern since the light doping necessary to produce antiferromagnetism in the alloys indicates that impurity effects can be important. An interesting difference in the interaction between superconductivity and magnetic ordering in the UPt_3 and URu_2Si_2 (see sec. 3.3) compounds is that the order parameter in the latter is apparently unaffected by the superconducting transition, while it ceases to evolve below T_C in the former. The onset of superconductivity seems to interfere with magnetic ordering in UPt_3.

3.2 UBe_{13}

Inelastic polarized and unpolarized neutron scattering measurements on polycrystalline samples of UBe_{13} were made by Goldman et al [24]. The inelastic spectrum is characterized by a broad quasielastic response ($\Gamma = 13 \pm 2$ meV) at 10K which exhausts most of the bulk susceptibility at this temperature. As temperature increases, the linewidth grows, saturating at $\Gamma \approx 40$ meV above 200K [25]. This behavior is reminiscent of the temperature dependence of the linewidth in the Ce HFS, albeit on a significantly larger absolute scale. The Q-dependence of the inelastic scattering at an energy loss of 12 meV (2 meV energy resolution) was well described by the U 5f magnetic form factor.

More recently, Neumann et al [26] have used very coarse energy resolution to reexamine the Q-dependence of the magnetic scattering from a polycrystalline sample with polarized neutrons. With the spectrometer set for elastic scattering, the wide energy window is expected to effectively integrate over the bulk of the magnetic response. However, it should be pointed out that this measurement is quite difficult and susceptible to error. Although not explicitly stated in the paper, it is assumed that the data were corrected for the finite flipping ratio of the spectrometer to account for feedthrough of the nuclear scattering and, the contribution of higher order scattering for which the flipping efficiency is probably much lower. Their data show a weak enhancement of the scattering at a momentum transfer, $Q=1\text{Å}^{-1}$, consistent with a maximum in the magnetic diffuse scattering near the (111) reciprocal lattice position. This can result from antiferromagnetic correlations between the U ions in the cubic unit cell. However, this observation must be reconciled with the featureless Q-dependence observed in the previous polycrystalline measurement.

One possible explanation of this discrepency is that there are additional spin fluctuation modes in UBe_{13} at some small energy scale, which are strongly $\vec{q}$-dependent. Since most of the bulk susceptibility is exhausted by the high frequency response, the spectral weight carried by this narrow energy feature must be quite small. However, its contribution to the magnetic scattering at certain points in reciprocal space can be enhanced by strong $\vec{q}$-dependence. Such a narrow feature has in fact been observed in single crystal measurements using unpolarized neutrons by Mook et al [27]. The quasielastic half-width of the response is less than 3 meV, and the magnetic scattering is a maximum at (111).

Preliminary polarized beam measurements on single crystals [28] of UBe_{13} are not inconsistent with this explanation. It then follows that the modulation was not observed in reference 24 because the Q-dependence in this case was measured at a finite (12 meV) energy transfer, with good resolution, so this low energy response lay far outside the region of measurement. Further polarized and unpolarized neutron scattering studies of single crystal specimens are in progress.

3.3 URu_2Si_2

One of the newest HFS, URu_2Si_2 has generated a great deal of excitement because it was the first such system in which a magnetically ordered state ($T_N = 17.5$K) and superconductivity ($T_C \approx 1.5$K) were found to coexist [29]. This phenomenon has been observed before in the rare earth ternary superconductors [30], but here the ordered moment is associated with

the rare earth ion, on its own sublattice, while the superconducting charge carriers are derived from other *constituents*. The effective interaction between the ordered moments and the conduction electrons is quite weak. In contrast, for URu_2Si_2, both the magnetic ordering and the superconductivity involve the U 5f electrons.

Walter et al [31] performed unpolarized neutron scattering measurements on a polycrystalline sample, and reported the existence of a gap in the magnetic excitation spectrum at low temperature (10K). Broholm et al [32], studying a single crystal, found that the transition at 17.5K was to an antiferromagnetic state with a very small ordered moment ($0.03 \pm 0.01\mu_B$) oriented along the tetragonal c-axis. The magnetic structure is characterized by ferromagnetic planes of U ions separated by c/2, which are antiferromagnetically coupled. No anomalous behavior of the order parameter was observed below the superconducting transition.

Inelastic neutron scattering measurements [32] found intense, well defined propagating magnetic excitations in the ordered phase. This behavior is in sharp contrast to the inelastic spectrum of U_2Zn_{17} below T_N, and uranium intermetallic systems in general [33]. No change in the damping or intensity of the excitations in URu_2Si_2 is observed in the superconducting phase.

In addition to the anomalies observed in the resistivity and susceptibility which are associated with the antiferromagnetic ordering at 17.5K, broader anomalies are observed at higher temperatures ($\approx$ 50K) [29]. In particular, the temperature dependence of the resistivity, and structure in the susceptibility measurements are similar to UPt_3. It seems plausible that magnetic correlations analagous to those found in UPt_3, near 30K, may be observed in URu_2Si_2 for $T_N < T < 50K$, and this should be investigated.

3.4 U_2Zn_{17}

The magnetic structure of U_2Zn_{17} was determined from neutron diffraction measurements by Cox et al [34]. The transition at 10K is to an antiferromagnetic state in which the two U ions in the rhombohedral unit cell are antiferromagnetically coupled. The magnetic unit cell is the same as the chemical unit cell. The moment direction lies in the basal plane, but could not be determined uniquely from the powder data. In addition, as seems to be the case for all magnerically ordered HFS, the ordered moment ($0.8 \pm 0.1\mu_B$) is substantially reduced from the paramagnetic moment of $2.25\mu_B$/U ion.

Inelastic measurements on single crystal samples have been reported by Broholm et al [35]. No sharp excitations are observed in the ordered phase. Rather, a single ridge in $S(\vec{q},\omega)$, parallel to the ω-axis, is seen, which broadens as the temperature is increased beyond T_N. Fits to the inelastic data using a model which incorporates both the single ion Kondo interaction between the f-electrons and the conduction electrons, as well as an RKKY interaction, were performed. Within the framework of this model, both interactions display temperature dependence, and it was concluded that it is the temperature dependence of the RKKY exchange which drives the magnetic transition in accord with the prediction of recent theories [1].

4. Discussion

The onset of magnetic correlations between the f-electrons in HFS at low temperature has been firmly established by neutron scattering measurements. As the number of studies on single crystal samples increases, it seems clear that this is a universal feature of the HFS regardless of the low temperature ground state of the particular system (eg. paramagnetic ($CeCu_6$), magnetic (U_2Zn_{17}), superconducting (UBe_{13}), or both magnetic and superconducting (URu_2Si_2, UPt_3)).

For those HFS that order magnetically, the ordered moment is significantly smaller ($< 1\mu_B$) than the free ion moment calculated from high temperature susceptibility measurements ($> 2\mu_B$) for both the Ce and U based systems. In particular, in both URu_2Si_2 and UPt_3, the ordered antiferromagnetic state carries a very small moment ($\approx 0.02\mu_B$) which coexists with the superconductivity. The bulk of the moment in UPt_3 is found in antiferromagnetic spin fluctuations, at higher frequency, of an entirely different character.

Only those systems shown by neutron scattering measurements to exhibit well developed magnetic correlations at low temperature have been discussed in this review. However, it is worthwhile at this point to mention other HFS which should be investigated in the near future. Both $CeCu_2Si_2$ and $CeAl_3$ exhibit anomalies, in for example low temperature magnetoresistance measurements [36], which are characteristic of the onset of coherence between the f-electrons. In $CeCu_2Si_2$, this behavior is sample dependent, and good single crystals of $CeAl_3$ have not yet been produced. The alloy, $U_{1-x}Th_xBe_{13}$ has attracted a great deal of attention because of the appearance of a second peak in specific heat [37] and ultrasound [38] measurements below the superconducting transition for x=2-4 at%. Batlogg et al [38] originally proposed a spin density wave (SDW) interpretation of this feature, and Machida and Kato [4] have recently studied the relationship between such a SDW and superconductivity in $U_{1-x}Th_xBe_{13}$. NMR and muon-spin relaxation measurements [39] indicate that the ordered moment in this system must be less than $10^{-2} - 10^{-4}\mu_B$, and no evidence of an ordered moment has been found from neutron scattering [40]. However, in light of the small moments found in URu_2Si_2 and UPt_3, further study of $U_{1-x}Th_xBe_{13}$ is in order.

Finally, recent measurements on UPt_3 have found a strong interaction between the antiferromagnetic order and superconductivity. This is perhaps not unexpected since we know from studies on the diluted systems [41] that there is a strong competition between magnetic order and superconductivity to begin with. Several papers [42] in the last year have established a connection between the symmetry of the superconducting ground state and the experimental observation of antiferromagnetic spin fluctuations. Further theoretical work on the interaction

between magnetic order and superconductivity is surely forthcoming.

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